

Amendments to the Specification:

Please replace the paragraph on page 2, lines 9-16, with the following amended paragraph:

Also, proposed (JP-A-60-28826) are composites each comprising a porous supporting body of a metal, an inorganic or polymer substance and a thin membrane of a cage type zeolite integrated in one surface. Among them, those having high affinity for a gel substance are proposed as especially preferable ones for the supporting body to be used and practically, it is proposed to use No. 7930 product produced by Corning Glass Works, generally called Vycor glass, as an especially preferable one.

Please replace the paragraphs on page 2, line 24 - page 3, line 14, with the following amended paragraphs:

Further, another method proposed (Japanese Patent Laid-Open No. 6-32610) relates to a method for production of an A-type or faujasite type zeolite membrane using a substrate of a substance mainly containing silicon oxide. The method aims to solve the problem of inferior adhesion strength of a zeolite membrane to a substrate, wherein a zeolite membrane is used as a substrate itself and the substrate surface is made to be a zeolite membrane owing to its constitution, thereby the synthesis and the adhesion can simultaneously be carried out to simplify the processes. To be practically practical, a substrate made of borosilicate glass, quartz glass, silica-alumina, mullite or the like is proposed.

Further, there is another proposal (JP-A-9-173799) which relates to a production method of a carrier zeolite membrane, and a membrane as the carrier, comprises including an inorganic, organic, or mixed substance selected from the group consisting of a ceramic

substance basically containing alumina, zirconia, or titanium oxide; a metal; carbon; silica; a zeolite; a clay; and a polymer.

Please replace the paragraph on page 3, line 23 - page 4, line 5, with the following amended paragraph:

That is, as shown in Figure 16, the thermal expansion coefficient of a zeolite shows a rather complicated behavior; at a temperature to around 200°C, it is extremely low but it becomes a negative coefficient value at a temperature ~~further~~ higher than that. Hence, if a zeolite membrane is to be used at a temperature exceeding 200°C, the thermal expansion coefficient difference becomes extremely high between a substrate, for example, an alumina-based substrate and the membrane, resulting in cracking of the zeolite membrane owing to the thermal stress.

Please replace the paragraph on page 4, line 22 - page 5, line 14, with the following amended paragraph:

Further, the following are proposed as examples of those having double layer ~~structure~~ structures of a substrate and a zeolite membrane: asymmetric membranes (JP-A-7-505333) each comprising a macroporous layer formed practically only from a molecular sieve crystal with a prescribed thickness and an upper layer for molecular separation having a prescribed thickness and a prescribed effective diameter of fine pores and formed practically only from the molecular sieve crystal of the same type as that of the material of the macroporous layer; a structure (JP-K-11-511685) composed of three layers, a carrier, an intermediate layer, and an upper layer and in which the intermediate layer and the upper layer contain prescribed crystalline molecular sieves; and a zeolite composite membrane (International Laid-open No. WO 00/23378) produced by forming a zeolite membrane containing a template on a zeolite

shaped body containing a template and then calcining to form the membrane and simultaneously remove the template. These membranes and structure are respectively excellent in the properties; the capability of precisely adjusting the size of the fine pores and the capability of effectively preventing occurrence of cracking.

Please replace the heading on page 8, line 4, with the following amended heading:

DISCLOSURE SUMMARY OF THE INVENTION

Please replace the paragraphs on page 8, line 9 - page 16, line 19, with the following amended paragraphs:

[1] — A In one aspect of the invention, there is provided a porous zeolite shaped body of a zeolite, characterized in that a porous zeolite shaped body is made of a completely crystallized zeolite composed of tetrapropylammonium ion (TPA) and silica sol in a mole ratio (TPA/SiO₂) of 0.015 to 0.08.

[2] — A In another aspect of the invention, there is provided a porous zeolite shaped body of a zeolite, characterized in that a porous zeolite shaped body is made of a zeolite still under the crystallization and composed of tetrapropylammonium ion (TPA) and silica sol in a mole ratio (TPA/SiO₂) of 0.02 to 0.12.

[3] — A In another aspect of the invention, there is provided a zeolite intermediate body, which includes one of the zeolite shaped bodies discussed above containing body, characterized in that the zeolite shaped body as described in [1] or [2] contains further a template, and a template-containing zeolite membrane having a composition the same as or similar to that of the shaped body is formed on the zeolite shaped body thereon.

[4] — A In another aspect of the invention, there is provided a zeolite layered composite comprising said a zeolite shaped body and said a zeolite membrane layered

thereon, characterized in that the composite is produced by removing said the template from said the zeolite shaped body and said the template-containing zeolite membrane by calcining the zeolite layered intermediate body as described in [3].

[5] — A In another aspect of the invention, there is provided a method for producing a zeolite layered composite characterized by comprising layering a template-containing zeolite membrane having a composition the same as or similar to that of a zeolite shaped body of a completely crystallized zeolite composed of tetrapropylammonium ion (TPA) and silica sol in a mole ratio (TPA/SiO₂) of 0.015 to 0.08 and containing a template therein on said the zeolite shaped body[[,]]; and simultaneously removing the template from said the zeolite membrane and said the zeolite shaped body by calcining the resulting layered product to obtain the zeolite layered composite comprising said including the zeolite shaped body and said with the zeolite membrane layered thereon.

[6] — A In another aspect of the invention, there is provided a method for producing a zeolite layered composite characterized by comprising layering a template-containing zeolite membrane having a composition the same as or similar to that of a zeolite shaped body of a zeolite still under crystallization and composed of tetrapropylammonium ion (TPA) and silica sol in a mixing ratio (TPA/SiO₂) of 0.02 to 0.12 by mole and containing a template therein on said the zeolite shaped body[[,]]; and simultaneously removing the template from said the zeolite membrane and said the zeolite shaped body by calcining the resulting layered product to obtain the zeolite layered composite comprising said including the zeolite shaped body and said with the zeolite membrane layered thereon.

[7] — A In another aspect of the invention, there is provided a porous zeolite shaped body of a zeolite, characterized in that the porous zeolite shaped body which has an average particle diameter of 1.0 μm or larger, a bending strength of 1.5 MPa or higher, and a difference in pressure between a feed side and a permeation side of 1.0 atmospheric pressure

or lower at 10 ml/cm² · min of helium gas permeation flux when a thickness of the porous zeolite shaped body is adjusted to be 1.8 mm.

[8]—A In another aspect of the invention, there is provided a zeolite layered intermediate body, characterized in that wherein the zeolite shaped body as described in [7] contains in the immediately preceding paragraph further comprises a template, and further comprising a template-containing zeolite membrane having a composition the same as or similar to that of the zeolite shaped body is layered thereon on the zeolite shaped body.

[9]—A In another aspect of the invention, there is provided a zeolite layered composite comprising said a zeolite shaped body and said a zeolite membrane layered thereon, characterized in that the zeolite layered composite is being formed by removing said the template from said the zeolite shaped body and said the template-containing zeolite membrane by calcining the zeolite layered intermediate body as described in [8].

[10]—A In another aspect of the invention, there is provided a method for producing a zeolite shaped body, comprising characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution and tetrapropylammonium bromide (TPABr) to silica sol in such a manner that mixing ratios [TPAOH/(TPAOH + TPABr) and TPABr/(TPAOH + TPABr)] of tetrapropylammonium hydroxide (TPAOH) and tetrapropylammonium bromide (TPABr) to a total amount of tetrapropylammonium ion (TPA) become 0 to 99% by mole and 1 to 100% by mole, respectively, to prepare a solution[[,]]; drying the prepared solution by kneading the solution[[,]]; shaping the obtained dried gel[[,]]; and subjecting the shaped body to a crystallization treatment.

[11]—A method for producing a zeolite shaped body, characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol to prepare a solution, spraying the prepared solution

to dry, shaping the obtained dried gel, and subjecting the shaped body to crystallization treatment.

[12]—A method for producing a zeolite layered intermediate body, characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution and tetrapropylammonium bromide (TPABr) to silica sol in such a manner that mixing ratios [TPAOH/(TPAOH + TPABr) and TPABr/(TPAOH + TPABr)] of tetrapropylammonium hydroxide (TPAOH) and tetrapropylammonium bromide (TPABr) to a total amount of tetrapropylammonium ion (TPA) become 0 to 99% by mole and 1 to 100% by mole, respectively to prepare a solution, drying the prepared solution by kneading the solution, shaping the obtained dried gel, subjecting the shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution having the same or similar composition as or to said prepared solution, and forming a template-containing zeolite membrane on said zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-containing zeolite membrane.

[13]—A method for producing a zeolite layered intermediate body, characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol, spraying thus prepared solution to dry, shaping the obtained dried gel, subjecting the shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-containing zeolite membrane.

[14]—A method for producing a zeolite layered composite, characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution and tetrapropylammonium bromide (TPABr) to silica sol in such a manner that the mole ratio of mixing ratios [TPAOH/(TPAOH + TPABr) and TPABr/(TPAOH + TPABr)] of tetrapropylammonium hydroxide (TPAOH) and tetrapropylammonium bromide (TPABr) to a total amount of tetrapropylammonium ion (TPA) become 0 to 99% and 1 to 100%, respectively to prepare a solution, drying the prepared solution by kneading the solution, shaping the obtained dried gel, subjecting the shaped body to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said solution, forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-containing zeolite membrane, and then calcining the layered body to simultaneously removing the template.

[15]—A method for producing a zeolite layered composite, characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol to prepare a solution, spraying thus prepared solution to dry, shaping the obtained dried gel, subjecting the shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution having the same or similar composition as or to that of said prepared solution, forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-containing zeolite membrane, and then calcining the layered body to simultaneously removing the template.

[16]—A In a further aspect of the present invention, there is provided a porous zeolite shaped body of a zeolite, characterized in that wherein area of parts (sound parts) where respective particles are clearly observed by grain boundary fracture among particles composing the zeolite shaped body in microstructure observation of the fractured surface of the shaped body occupies 70% or more in the entire area of the fractured surface.

[17]—A In a further aspect of the present invention, there is provided a zeolite layered intermediate body, characterized in that wherein the zeolite shaped body as described in [16] the preceding paragraph contains a template, and a template-containing zeolite membrane having a composition the same as or similar to that of the shaped body is formed on the shaped body.

[18]—A In a further aspect of the present invention, there is provided a zeolite layered composite comprising a zeolite shaped body and a zeolite membrane formed thereon, characterized in that wherein the zeolite layered composite is produced by removing said template from said zeolite shaped body and said template-containing zeolite membrane by calcining the zeolite layered intermediate body as described in [17] the preceding paragraph.

[19]—A method for producing a zeolite shaped body characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO₂) of tetrapropylammonium ion (TPA) to the silica sol becomes 0.015 to 0.08 by mole to prepare a solution, drying the prepared solution by kneading the solution, wet pulverizing the obtained dried gel, drying the obtained slurry by spraying the slurry, shaping the obtained dried granular substance, and subjecting thus shaped body to crystallization treatment.

[20]—A method for producing a zeolite shaped body, characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO₂) of

tetrapropylammonium ion (TPA) to the silica sol becomes 0.015 to 0.08 by mole to prepare a solution, spraying thus prepared solution to dry, shaping the obtained dried gel, and subjecting thus shaped body to crystallization treatment.

[21]—A method for producing a zeolite layered intermediate body, characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO₂) of tetrapropylammonium ion (TPA) to the silica sol becomes 0.015 to 0.08 by mole to prepare a solution, drying the prepared solution by kneading the solution, wet pulverizing the obtained dried gel, spraying the obtained slurry to dry, shaping the obtained dried granular substance, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising the zeolite shaped body and the template-containing zeolite membrane.

[22]—A method for producing a zeolite layered intermediate body characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO₂) of tetrapropylammonium ion (TPA) to the silica sol becomes 0.015 to 0.08 by mole to prepare a solution, spraying thus prepared solution to dry, shaping the obtained dried gel, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution having the same or similar composition as or to that of said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to

produce a layered body comprising the zeolite shaped body and the template-containing zeolite membrane.

[23]—A method for producing a zeolite layered composite, characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such manner that a mixing ratio (TPA/SiO₂) of tetrapropylammonium ion (TPA) to the silica sol becomes 0.015 to 0.08 by mole to prepare a solution, drying the prepared solution by kneading the solution, wet pulverizing the obtained dried gel, drying the obtained slurry by spraying the slurry, shaping the obtained dried granular substance, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution having the same or similar composition as or to that of said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising the zeolite shaped body and the template-containing zeolite membrane, and then simultaneously removing the template by calcining the layered body.

[24]—A method for producing a zeolite layered composite, characterized by in accordance with another aspect of the present invention comprises adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such manner that a mixing ratio (TPA/SiO₂) of tetrapropylammonium ion (TPA) to the silica sol becomes 0.015 to 0.08 by mole to prepare a solution, spraying thus prepared solution to dry, shaping the obtained dried gel, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said solution, forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising the zeolite shaped body and the template-containing zeolite membrane, and then

simultaneously removing the template by calcining the layered body.

Please replace the heading on page 19, line 13, with the following amended heading:

~~BEST MODE FOR CARRYING OUT~~ DETAILED DESCRIPTION OF THE INVENTION

Please replace the paragraphs on page 19, line 14 - page 20, line 10, with the following amended paragraphs:

Since a zeolite shaped body of the invention is to be effectively used as a substrate in form of a zeolite layered composite by layering or forming a zeolite membrane thereon for a gas separation membrane of a molecular sieve membrane and a pervaporation membrane, it is required to prevent cracking in the zeolite to be layered or formed thereon. For that, a zeolite shaped body of the invention is ~~preferable to be~~ preferably a porous zeolite composed of particles of a zeolite with the composition which is the same as or similar to that of the zeolite membrane to be layered thereon in the case where it is used for obtaining a zeolite layered composite by layering the zeolite membrane thereon.

Especially, in the case where a zeolite layered composite is formed using a template, in consideration of the fact that the thermal expansion behavior of a template-containing zeolite membrane is extremely different from that of a zeolite membrane containing no template as shown in Figure 17, only using a substrate (for example, quartz glass and the like) with the thermal expansion coefficient approximate to that of the zeolite membrane is insufficient to solve the thermal expansion difference at the time of calcining at about 500°C for removal of the template and consequently cracking takes place in the zeolite membrane, and therefore, a zeolite shaped body of the invention is preferably a porous zeolite with the composition which is the same as or similar to that of the zeolite membrane in the entire respect with the composition including the template.

Please replace the paragraph on page 32, lines 11 - 16, with the following amended paragraph:

Further, a zeolite layered composite of the second invention is characterized by comprising a zeolite shaped body with a zeolite membrane layered on the shaped body and being produced by removing the template from the zeolite shaped body and the zeolite-containing zeolite membrane by calcining said zeolite layered intermediate body.

Please replace the paragraph on page 36, lines 3 - 9, with the following amended paragraph:

Then, the dried gel is shaped by properly forming the gel in a prescribed shape by a uniaxial pressing with a die (the total pressure 1,000 kgf) and then carrying out cold isostatic pressing to obtain a dried gel shaped body. At this time, the pressure of the cold isostatic pressing is preferable to be preferably adjusted within a range of 700 to 7,000 kgf/cm² so as to make the resulting dried gel shaped body to have a desired density.

Please replace the paragraph on page 52, lines 17 - 21, with the following amended paragraph:

In the examples of the third invention, the TPA/SiO₂ mole ratio is adjusted to be 0.04 at which the bending strength becomes the maximum and as a tetrapropylammonium ion (TPA) source, a tetrapropylammonium hydroxide (TPAOH) solution is used to produced produce the solution.

Please replace the paragraph on page 69, lines 10 - 24, with the following amended paragraph:

A Teflon beaker of 200 ml capacity was charged with about 30% by weight of silica

sol (trade name: Snowtex S, produced by Corporation Nissan Chemical) and a 10% tetrapropylammonium hydroxide solution (produced by Wako Pure Chemical Industries, Ltd.) and the mixing ratio (TPA/SiO₂) of TPA (tetrapropylammonium ion) and SiO₂ was separately adjusted as shown in Table 2 [in mole ratio] and the resulting each solution was stirred at room temperature for 30 minutes by a magnetic stirrer and after that, while ~~being~~ heating at 80°C, each solution was further continuously stirred and kneaded to evaporate water and to obtain a colorless dried gel with 10% by weight or lower water content. The obtained each dried gel was subjected to x-ray diffraction to investigate the crystal structure to find it was amorphous.

Please replace the paragraph on page 70, line 17 - page 71, line 6, with the following amended paragraph:

A Teflon beaker of 200 ml capacity was charged with about 30% by weight of silica sol (trade name: Snowtex S, produced by Corporation Nissan Chemical) and a 10% tetrapropylammonium hydroxide (TPAOH) solution (produced by Wako Pure Chemical Industries, Ltd.) while the mixing ratio (TPA/SiO₂) of TPA (tetrapropylammonium ion) of the TPAOH solution and silica sol ~~being~~was adjusted to be 0.04 by mole ratio and the resulting solution was stirred at room temperature for 30 minutes by a magnetic stirrer to obtain a mixed solution of tetrapropylammonium hydroxide (TPAOH) and silica sol for a spray drier. The mixed solution was dried by a spray drier apparatus (trade name: Valvis Mini Spray GA 32 model manufactured by Yamato Science Co., Ltd.) in conditions of 1 kgf/cm² spraying air pressure, 0.4 m³/min dried air flow rate, 3 ml/min solution feeding rate, and 180°C blowing temperature to obtain a dried gel. The obtained dried gel was subjected to x-ray diffraction to investigate the crystal structure to find it was amorphous.

Please replace the paragraph on page 73, lines 1 - 18, with the following amended paragraph:

Table 4 collectively shows the results of the measurement for the average particle diameter (mm) of the microstructure of the fractured surface of each zeolite shaped body obtained in examples 14 to 20 and the comparative example 16, the four-point bending strength (MPa) and pressure loss (atm) of each zeolite shaped body obtained in examples 14 to 20 and the comparative example 16. According to Table 4, the zeolite shaped bodies obtained in examples 14 to 20 had practically sufficiently large average particle diameter and high bending strength as compared with those of the zeolite shaped body obtained in the comparative example 16 and their pressure loss was found to be extremely low. Consequently, if a zeolite layered composite comprising a shaped body (a substrate) with such an extremely low pressure loss just like the zeolite shaped bodies obtained in the examples 14 to 20 and a zeolite membrane free of defects such as cracks and layered or formed thereon is used as a gas separation membrane of a molecular sieve membrane and a pervaporation membrane, the composite can be a highly functional and useful material with a high flux.